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Magnetocaloric effect description and principle of operation

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Abstract: The following article is an explanation of the phenomenon of magnetocaloric phenomenon, which describes the principle of refrigerants in modern magnetic refrigerators. The magnetocaloric effect occurs over a wide temperature range from low to high temperatures. MCE is observed in various materials such as transition metals, rare earth elements, Laves phases, manganites, $La(Fe,Si)_{13}$ alloys, full and half Heusler alloys and MM'X alloys. The principle of magnetic chillers is also described.

Key words: magnetocaloric effect, structure, MM'X alloys, magnetocaloric materials, magnetic coolers, AMR

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1. Introduction

The magnetocaloric effect (MCE) is described as an adiabatic change in temperature ΔT_{ad}. This phenomenon consists of two stages. The first stage relates to heating during magnetisation, which is the fact that when an external magnetic field is applied to a material, the magnetic moments of the spins order themselves, thus reducing the magnetic entropy of the system and increasing the temperature of the material. The second stage refers to the cooling during demagnetisation, which is related to the fact that when the external magnetic field is removed the magnetic moments of the spins are reoriented, which has the effect of lowering the temperature of the material as the domains absorb thermal energy to re-disorder (Pecharsky and Gschneidner, 1999).

The history of the magnetocaloric effect dates back to 1860, when W. Thomson (Thomson. 1860) described the first theoretical prediction of the occurrence of the phenomenon. The next reference to MCE was described by E. Warburg (Warburg 1881) in 1881, discovering the phenomenon as heat release in iron under the influence of an external magnetic field. In 1917, P. Weiss and A. Piccard (Weiss and Piccard, 1979) discovered the magnetocaloric effect. They observed a change in the temperature of pure Ni under the influence of an external magnetic field. The practical application and mechanism of the effect to achieve very low temperatures in the demagnetisation of paramagnetics was studied by the non-magnetic P. Debye in 1926 (Debye 1926) and W. Giauque in 1927 (Giaque, 1927). However, it was in 1997 that V. Pecharsky and K. Gschneidner Jr (Pecharsky and Gschneidner, 1997) discovered the giant magnetocaloric effect in $Gd_5Ge_2Si_2$-type alloys, which led to an avalanche of research interest in the topic of the magnetocaloric effect.

2. MCE in different temperature range

The occurrence of the magnetocaloric effect varies depending on the temperature range. The phenomenon occurs in a temperature range such as low temperatures, room temperature and high temperatures, as well as a phase transition range. This effect is also sensitive to material properties. Each temperature range requires the material selection to be adjusted in order to effectively optimise the MCE effect for use in cooling technology.

2.1. Low temperature range

Materials such as pure lanthanide group metals (e.g. Gd and its alloys) exhibit magnetocaloric phenomena at low temperatures (close to absolute zero). The mechanism of the phenomenon in this temperature range is based on the fact that the magnetic moments of the atoms are disordered, and when an external magnetic field is applied, an equalisation of the

moments is observed, the entropy decreases and the temperature of the material increases (Pecharsky and Gschneidner, 1999).

2.2. Room temperature range

The prototype material for this temperature range was Gd. This material tidies up ferromagnetically at temperatures below 294K. This change is influenced by the purity of the element. MCE is also observed in transition metal alloys, as well as their oxides. The mechanism of MCE is strong near the Curie temperature (T_C), where the material changes from a ferromagnetic to a paramagnetic state. This change can be used for magnetic cooling (Pecharsky and Gschneidner, 1999).

2.3. High temperature range

Some high-temperature alloys such as $La(Fe, Si)_{13}$, for example, exhibit the phenomenon in the high temperature range. They are developed in such a way as to maintain a significant MCE. The mechanism of the effect in this temperature range is based on the behaviour of magnetic moments, the alignment of which is com- plicated by the increased temperature, but some materials still exhibit significant MCE due to the trans-formation of the magnetic structure (Pecharsky and Gschneidner, 1999).

2.4. Phase transition range

The magnetocaloric effect is pronounced near phase transitions such as martensitic or structural transitions. These transitions involve changes in magnetic ordering or structure. The mechanism of the effect in this temperature range is related to the arrangement of the magnetic moments, which are drastically altered by the influence of an external magnetic field that leads to visible changes in the temperature of the material (Pecharsky and Gschneidner, 1999).

3. Materials that demonstrating MCE

The magnetocaloric effect is observed for all magnetic materials. An increase in the MCE value is associated with an increase in the applied magnetic field. Several groups of magnetocaloric materials can be distinguished, such as 3d transition metals (and their alloys), rare earth elements (and their alloys), Laves phases, manganites, $La(Fe,Si)_{13}$-type alloys, Heusler alloys and half Heusler and MM'X-type alloys.

The magnetocaloric phenomenon is described by the values of adiabatic temperature change (ΔT_{ad}) and magnetic entropy variation (ΔS_M), which are correlated with magnetic field strength (H), magnetisation (M), absolute temperature (T) and heat capacity (C) at constant pressure. Using Maxwell's basic relation, the value of ΔS_M can be described by the relation (Pecharsky and Gschneidner, 1999):

$$\Delta S_M(T, \Delta H) = \int_{H_1}^{H_2} \left(\frac{\partial M(T,H)}{\partial T}\right)_H \quad (1)$$

and the measure of MCE can be described by the equation (Pecharsky and Gschneidner, 1999):

$$\Delta T_{ad}(T, \Delta H) = -\int_{H_1}^{H_2} \left(\frac{T}{C(T,H)}\right)_H \left(\frac{\partial M(T,H)}{\partial T}\right)_H dH \quad (2)$$

Equations (1) and (2) are of fundamental non-negligible importance for understanding the behaviour of MCE in solids and also serve as a basis in the search for new magnetocaloric materials.

One of the basic criteria for the selection of magnetocaloric materials is maximum efficiency with the lowest possible magnetic field variation. Such conditions are fulfilled for paramagnetic materials at very low temperatures. Another selection criterion is the use of magnetic materials with high spin density and high magnetic moments. Materials that exhibit second-order magnetic transformation have a wide temperature range of magnetic transformation. This is an important criterion from an application point of view, as this temperature affects the value of the cooling capacity (RC). This quantity determines the maximum energy that magnetocaloric materials will be able to exchange (Gschneidner et al., 2005; Mayer, 2011).

3.1. 3d transition metals and their alloys

The 3d materials in which MCE has been measured include: Fe, Co, as well as Ni (Gschneidner and Pecharsky, 2000). The maximum ΔT_{ad} values for iron are 2K at an external magnetic field of 0.8T, and also 5.2K at a field of 3T. The ΔT_{ad} value for cobalt was 3.25K at 2.32T, and for nickel 0.57K at 2.5T. The behaviour of ΔT_{ad} with respect to the magnetic field is not linear. Considering 3d transition metal-based alloys such as: $Fe_{0.9357}Si_{0.0643}$ (Hirschler and Rocker, 1966) the MCE value reaches a maximum of 5K at a magnetic field of 3T. For the alloy $Mn(As_{1-x}P_x)$ (Gschneidner and Pecharsky, 2000) the value of MCE at an external magnetic field of 7T is 1.3K. The value of the magnetocaloric phenomenon for transition metals shows complex dependencies as a function of temperature and magnetic field (Gschneidner and Pecharsky, 2000).

3.2. Rare earth elemenets and their alloys

Rare earth elements are part of the lantan group. They include elements such as Dy, Er, Tb, Nd, La and Gd. The latter (Gd) has been one of the most extensively studied rare earth elements for the occurrence of the magnetocaloric phenomenon. This element has strong ferromagnetic properties in the temperature range from liquid helium (He) to 294K. Thanks to research on the $Gd_5Ge_2Si_2$ alloy (Tripathy et al., 2006) and the discovery of giant MCE, this element has become a reference for new materials exhibiting a magnetocaloric effect at ambient temperature. Of all the primes, compounds that were based on Gd were extensively studied because gadolinium has the highest magnetic moment, as well as the highest effective exchange coupling at ambient temperature. Another reason was that Gd has no orbital moment in its ground state,

which facilitated the understanding of the magnetothermal behaviour occurring in compounds containing this element (Tishin and Spichkin, 2003).

3.3. Lavesa phases

The group of these compounds is described using the general formula AB_2. A distinction is made between 3 basic groups of Laves phases such as regular C15, hexagonal type C14 and hexagonal type C36. Gschneidner et al (Gschneidner et al., 1994) conducted research on alloys such as RCo_2, RAl_2 or RNi_2. They placed particular emphasis on the RCo_2 alloy (where R=Dy, Ho, Er), in which a paramagnetic-ferromagnetic transition of the first type was observed, while others show a ferromagnetic transition of the second type. Experimental and theoretical studies have been conducted on RAl_2 alloys, as well as RNi_2 (Gschneidner et al., 1994).

3.4. Manganites

Manganites are ferromagnetic materials with a perovskite structure of the type $R_{1-x}M_xMnO_3$ (where R=La, Nd, Pr; M=Ca, Sr, Ba). This group of materials is used in magnetic cooling in the temperature range 100 to 375K. They have excellent magnetocaloric properties and have a low processing cost. Ongoing research on this group of alloys has shown a high and reversible magnetocaloric effect (Phan and Yu, 2007).

3.5. La(Fe,Si)13 type alloys

This group of alloys is characterised by the presence of a metamagnetic phase transition of the first kind in the vicinity of the Curie temperature (T_C), confirmed by magnetic studies. In these alloys, the magnetic state changes from paramagnetic to ferromagnetic. The metamagnetic wandering electron phase transition (IEM) shows an increase in the transition field with temperature (Gębara and Pawlik, 2017).

3.4. Heusler and half-Heusler alloys

Full Heusler alloys are described by the general formula X2YZ, and half Heusler alloys are described by the formula XYZ (where X,Y - atoms from the side group, Z - atom from the main group). The group of these compounds crystallise in two phases: hexagonal and rhombic. Despite the fact that they consist exclusively of paramagnetic elements, and become ferromagnets after appropriate treatment, they are characterised by remarkable magnetic properties. These materials are promising candidates as environmentally friendly and low-cost thermoelectric materials (Tishin and Spichkin, 2003; Potter, 1934).

3.5. MM'X type alloys

The group of compounds MM'X (where M, M' - transition metals, X - element from the main group) are characterised by ferromagnetic martensitic transformation of the first kind (FMMT). In FMMT, structural martensitic transformation is coupled with magnetic transformation, showing magneto-structural coupling. Studies show that the magnetostructural coupling, as well as the large magnetisation difference (ΔM) between the austenite and martensite phases, have a strong influence on the magnetocaloric effect. With decreasing temperature, the crystal structure changes from a high-temperature hexagonal Ni_2In type to a low-temperature rhombic NiTiSi type. Phases occurring in MM'X alloys are of great interest due to their giant magnetoresistance and giant MCE, which refers to the cooling or heating of the material under the influence of an applied external magnetic field (Liu et al. 2013).

4. MnCoGe alloys

MnCoGe alloys are the most common compound of the MM'X alloy group. They are characterised by the occurrence of two crystalline phases such as: low-temperature rhombohedral ($a\neq b\neq c$, $\alpha=\beta=\gamma=90^o$, cell shape - cuboid) with space group P_{nma} of NiTiSi type (Hahn 2002) and high-temperature hexagonal ($a=b\neq c$, $\alpha=\beta=90^o$, $\gamma=120^o$, cell shape - prism with square base) with space group $P6_{3/mmc}$ of Ni_2In type (Garcia et al. 2020). The martensitic transformation of the MnCoGe alloy occurs at a temperature of about 420K from a hexagonal to a rhombic phase. The Curie temperature (T_C) of the rhombohedral martensite is close to 355K and that of the hexagonal austenite around 276K. Due to the much higher transformation temperature of the martensite, no magnetostructural coupling is observed in the MnCoGe alloy (Bazela et al., 1976).

5. Effect of additives on MnCoGe alloys

Current research is being conducted into the effects of additives on the structure and magnetic properties of the MnCoGe alloy. Additives are selected in such a way as to obtain the highest possible value of the magnetocaloric effect. Additions of elements such as B (Trung et al. 2010), Ni (Zhang et al., 2015; Trung et al., 2010; Zhang et al., 2015; Kutynia et al., 2022; Gębara and Kutynia, 2019; Kutynia et al., 2024; Mieszczyński et al., 2023; Zeng et al., 2014; Kuang et al., 2022; Kanomata et al., 1995; Gębara and Śniadecki, 2019; Qian et al., 2021; Kutynia and Gębara, 2021; Gębara and Śniadecki, 2019; Kutynia et al., 2023) at the expense of Mn. These additions alter the structure, which may be due to the difference in ionic radii between Mn and the ionic radii of the added elements. The pro-rated studies on the above additives show that these elements improve the magnetic properties to a greater or lesser extent.

After analysing the results, it was observed that the addition of boron affects the occurrence of a giant MCE with a peak of ΔS_M=47.3 J(kg K)$^{-1}$ and a magnetostructural phase transition of the first kind was revealed. Similar effects were obtained for additions of nickel, carbon or gallium.

Additions of iron, aluminum, vanadium or copper at the expense of manganese resulted in lowering the temperature of

the martensitic phase transition to that of the magnetic transition. This resulted in both transitions occurring at room temperature.

The addition of tungsten, or niobium, had little effect on the occurrence of a significant magnetocaloric effect. However, an improvement in properties is observed.

Studies conducted on zirconium or palladium additives have shown quite significant effects on improving magnetic properties. The most favorable results were obtained for the addition of palladium, as a significant effect was observed on increasing the value of the change in magnetic entropy (ΔS_M), which affects the occurrence of a large magnetocaloric effect.

The additives studied to improve properties can be used as potential cooling agents in a new generation of magnetic refrigerators.

6. Principle of magnetic refrigerators

A description of the active magnetic regenerator (AMR) cycle, as well as the principle of operation, is presented in (Zimm et al. 1993). This cycle uses a porous magnetic refrigerant cartridge that acts as both a coolant (producing cooling) and a fluid (water) regenerator that transfers heat. The water that flows through the AMR transfers heat to, and from, external heat exchangers.

The entire cycle begins in a demagnetized state in a zero magnetic field. When an external magnetic field is applied, each particle that is in the magnetic material heats up due to the magnetocaloric effect, which results in the formation of the final magnetization temperature profile of the magnetic material. The value of the adiabatic temperature change (ΔT_{ad}) is equal to the amount of heat generated by the heating of each particle, which is reduced by the effect of capacitance in the pores between the particles. Further down the cycle, water flows through the magnetic material from the cold area to the hot area. The water-cooled material lowers the temperature profile across the magnetic cartridge. The water heated by the magnetic cartridge reaches a temperature close to that of the sample at the hot end. When the water flow is stopped, the magnetic field is removed, resulting in cooling of the magnetic material by the MCE. The post-high cycle ends by forcing a flow of fluid at about 24ºC from the hot end of the magnetic cartridge to the cold end. The water is cooled by the magnetic cartridge. The fluid flows in at a temperature below 5ºC and removes heat from the cooler as it passes through the cold heat exchanger (Pecharsky and Gschneidner, 1999).

6. Conclusion

In summary, current research on the magnetocaloric effect (MCE) is focused on the potential use of the phenomenon for cooling at room temperature. Magnetocaloric materials can be used for practical application as active magnetic regenerators in a new generation of magnetic refrigerators, which are expected to replace classic refrigerators in the future. An additional advantage of the new refrigerators is the efficiency achieved, which reached up to 60%, while conventional refrigerators reached about 40%. The lack of commercial application of magnetocaloric materials for the manufacture of magnetic refrigerators is due to the finding of low-cost materials with suitable magnetic properties and high magnetocaloric effect.

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